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**Proposal for Continuation of Research and
Semi-Annual Progress Report**

July 1, 1981 to December 31, 1981

on

**DENSITY MEASUREMENT IN AIR WITH
A SATURABLE ABSORBING SEED GAS**

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ABSTRACT

A proposal is made to continue a study of a method for making density measurements in a compressible flow by using off-resonance laser-induced fluorescence. The seed molecule chosen for study is the iodine molecule which is excited with the 514.5 nm line of the argon-ion laser whose output is frequency tuned, by as much as 3 GHz, relative to a strong iodine transition using an intracavity etalon. The theory which we recently developed to analyze the effect will be used in conjunction with two experiments being conducted to further study the method: an acoustic resonance tube in which controlled perturbations about a uniform state are produced, and a small supersonic jet in which the conditions of the flow vary widely from point to point.

SUMMARY OF WORK COMPLETED

A large portion of the effort in the last reporting period was devoted to the task of completing a report on the work that was done as part of J. C. McDaniel's Ph. D. thesis research. This report is listed as Ref. 1 in the list of references. In addition, we have recently completed the writing of a short paper which was submitted to the new letters section of The Physics of Fluids. A copy of this paper is included as an appendix to the present report. Our intent was to report on the approach we have been developing, to remove the effect of quenching by the use of off-resonance laser-induced fluorescence, in a letter with a relatively short publication time, and then follow it with a more extensive report on the theory in another publication. We are presently in the process of writing a paper on the theory and its application to the iodine molecule and plan to submit it for publication in Applied Optics in the very near future. Because a great deal of basic information related to the saturation properties of the iodine molecule was developed in our work, and this information is not available in the general literature, we plan to follow these publications with a paper discussing the saturation properties of iodine.

Part of our proposed work for the present year was to construct an acoustic resonance tube and determine whether one could obtain a modulated fluorescent signal which is in step with the pressure signal from a microphone placed in the tube. The purpose of this work was to study the application of McDaniel's theory of off-resonance laser-induced fluorescence in a controlled situation and determine the limits of sensitivity of the method and whether it can be applied to the study of gas dynamic flows which are the source of aerodynamic noise. This effort has been greatly aided by the addition to our group of Dr. Ulrich Ackermann, a visiting scientist from the DFVLR in Berlin, who will be spending the calendar year 1982 working with us on this problem. His home Institution is supporting his stay for the purpose of having him learn the method we are developing and to allow him to

familiarize himself with the laser equipment and experimental techniques required in the work. His ultimate objective is to work towards applying the method to a study of noise generation in a supersonic nozzle by localizing the sources of large density fluctuations in the turbulent part of the flow and then correlating them with the farfield sound measured by a microphone.

An acoustic resonance tube having a rectangular cross-section has been constructed by Dr. Ackermann and he has established that a sound pressure level of 150 db can be generated in the tube when driven at its resonance frequency by a speaker mounted at one end. An optical system has been set up to allow the insertion of the argon-ion laser beam into the resonance tube and to view the resulting fluorescence with a photomultiplier tube. The application of McDaniel's method requires a controlled detuning, by up to 3 GHz, of the laser frequency from the line center of the iodine absorption band used. Frequency tuning of the argon-ion laser is accomplished by changing the oven temperature of an etalon placed in the cavity of the argon-ion laser. We have been working with this system to learn how to use it effectively and to obtain a calibration curve of operating wavelength versus oven temperature, so that a scanning interferometer is not needed at all times to set the wavelength. We are nearly ready to search for the off-resonance laser-induced fluorescence signal in the acoustic resonance tube with our present setup. However, before the proper setting of the etalon was fully explored, Dr. Ackermann was able to obtain a sinusoidal fluorescent signal which was in phase with and at the same frequency as the electrical signal driving the speaker. Also, he was able to observe a fluorescent intensity pattern in the path of the laser beam which produce an image of the acoustic mode of oscillation of the resonance tube. These preliminary results lead us to believe that, after additional adjustments are made, we ought to be able to obtain some rather interesting results with the method and will be in a position to learn a great deal about the technique and its application.

Another part of the planned work for the present period was to further study the supersonic flow from a small underexpanded nozzle using iodine seeding of a nitrogen flow and the method of off-resonance excitation as developed by McDaniel. We have completed the construction of a small flow facility which can be placed on top of an optical table and allows the study of the flow from a small supersonic jet. One of the features of the chamber that encloses the jet is that its windows are set at Brewster's angle to the laser beam so that a smaller fraction of the laser light is reflected from the window surfaces than one would have with beams striking windows at normal incidence. A second feature of the facility is that provisions have been made to heat the reservoir gas and thereby change the vapor pressure of the iodine and the resulting iodine concentration in the nitrogen flow. In our preliminary studies, we have been able to reproduce McDaniel's findings with the new setup and we plan to continue his work in exploring the application of the method to the study of a supersonic flow.

In a closely related study on droplet evaporation using iodine as a fluorescent seed material, which is being supported by the Air Force, we conducted a study of the variation of the fluorescent intensity of iodine with changing iodine vapor pressure and nitrogen background pressure. The partial pressure of iodine was changed by raising and lowering the temperature of the iodine test cell and thereby altering the iodine vapor pressure. The observation that proved most striking was that a rather large hysteresis develops in the fluorescent intensity (or iodine partial pressure) as the iodine is taken through a heating and cooling cycle. It appears that the iodine passes from the solid to a gaseous phase during heating as expected, but on cooling down it exhibits a surprising lag in following the expected equilibrium vapor pressure curve. In other words the partial pressure of iodine is much greater than its equilibrium vapor pressure for a given temperature as the temperature is reduced. A possible explanation of this effect is that it may be difficult for the gas

phase iodine molecules to find the few solid nucleation sites that form on surfaces. The significance of this finding is that iodine condensation may be less of a problem in a supersonic flow than one would initially anticipate from its equilibrium vapor pressure curve.

PROPOSED WORK

We propose to continue working along the general lines described in the previous section. One of our principal objectives in the next year is to complete the writing of one or possibly two papers on topics found in McDaniel's thesis. Although McDaniel has finished his thesis, he will continue to be at Stanford for another year as a post-doctoral fellow and this will allow him to turn his attention to publishing the results of his work. Likewise, he will also be available to give advice on the continuing work as various problems arise.

With the completion of an acoustic resonance tube by Dr. Ackermann, we are in a position to turn our attention to using the device to explore the feasibility of detecting strong acoustics waves with the method of off-resonance laser-induced fluorescence. The study should answer a number of interesting questions with regard to the application of the method. Because signal strength is lost as the laser frequency is detuned from the iodine absorption line center, one would be interested in knowing how strong of an acoustic wave is needed for the wave to be detected by the method. The strength of the signal will also determine the upper limit on the frequency of the wave that can be detected. Both of these results will in a sense provide calibration information for the application of the method.

McDaniel's theory has not been studied in terms of small disturbances and we plan to look at his equations again to see if they suggest an optimum operating condition. His work focused on efforts to cancel the effect of quenching over a broad range of pressures. On the other hand, for small pressure changes one may find that rather different conclusions may be drawn. Here, we will have an excellent opportunity to compare the predictions of the theory with the results of our experiments.

The work with the small supersonic nozzle will be principally aimed at answer-

ing the question of whether iodine condensation affects the results obtained in a supersonic flow to a significant degree. Also it will allow us to further study the value of using the method in a situation where the pressure and temperature change by a large amount and consequently the quenching rate changes by a correspondingly large factor.

REFERENCE

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APPENDIX

(letter)

DENSITY MEASUREMENT IN COMPRESSIBLE FLOWS USING OFF-RESONANCE LASER-INDUCED FLUORESCENCE

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Measurement of molecular number density in compressible flows using laser-induced fluorescence is complicated by collisional quenching of the excited state. It is shown that by exciting the fluorescence off-resonance the signal becomes proportional to number density and independent of collisional effects. Quantitative measurement of density in an underexpanded jet of nitrogen is demonstrated using off-resonance fluorescence from iodine seed molecules irradiated with the 514.5 nm line of the argon-ion laser.

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Laser-induced fluorescence (LIF) is an attractive experimental technique for the nonintrusive measurement of the molecular number density at a point in a compressible flowfield^{1,2}. It provides density measurement capability with good spatial resolution and signal-to-noise ratio even at low concentrations of the fluorescing species. The primary drawback in using LIF to measure density is that the resulting signal is not simply proportional to density but, due to collisional quenching of the excited state, is also a strong function of pressure and temperature. Saturation of the absorbing transition has been proposed as a means for removing the quenching dependence of the LIF signal^{3,4}. However, complete saturation of a molecular transition under conditions of interest in gas dynamics has not been achieved^{5,6,7}.

In this letter we discuss an alternate approach to remove the quenching complication in LIF and enable quantitative density measurements to be made in a compressible flow. We refer to the method as off-resonance laser-induced fluorescence (ORLIF). In order to explore the concept we employed molecular iodine, seeded in the reservoir of a compressible flow of nitrogen, and induced the fluorescence with the 514.5 nm line of the argon-ion laser. We present here the essential elements of the theory and the experimental results which establish the validity and attractiveness of the ORLIF technique for nonintrusive density measurement.

Under the conditions of excitation by a narrow bandwidth laser and collection of the total broadband fluorescence the steady state solution of the rate equations for the fluorescent signal, neglecting saturation and radiative trapping effects, is⁷

$$S_F = \eta h\nu \frac{\Omega}{4\pi} V_c \left(\frac{4 \ln 2}{\pi} \right)^{1/2} \frac{A_{21}}{A_{21} + Q} \frac{I}{c} \frac{B_{12}}{\Delta\nu_d} V(D, B) f_1 f_\sigma N, \quad (1)$$

where the fluorescent signal, S_F , is defined as the number of photons of energy $h\nu$ per second striking the detector, collected at right angles to the laser beam by optics with collection efficiency, η , and solid angle, Ω , from a collection volume, V_c , within the flowfield. The spontaneous emission rate, A_{21} , and the collisional

quenching rate, Q , appear in the ratio $A_{21}/(A_{21} + Q)$ which is known as the Stern-Volmer⁸ or fluorescence efficiency factor. The laser intensity is denoted by I and the spec of light by ϵ . The absorbing transition is characterized by a line strength parameter, B_{12} , a Doppler width, $\Delta\nu_d$, and a Voigt line shape function, $V(D, B)$. The Voigt function is a convolution of the Lorentzian and Doppler distributions and accounts for the changing of the absorption line shape with varying thermodynamic conditions in the flowfield. This function contains a normalized detuning parameter

$$D = (4 \ln 2)^{1/2} \frac{\Delta\nu}{\Delta\nu_d}, \quad (2)$$

where $\Delta\nu$ is the difference between the laser frequency and the center frequency of the absorbing transition, and a normalized broadening parameter

$$B = (\ln 2)^{1/2} \frac{\Delta\nu_c}{\Delta\nu_d}, \quad (3)$$

where $\Delta\nu_c$ is the collision width. The fraction of seed molecules in the absorbing energy level is f_1 , the seeding fraction f_s is the ratio of the partial pressure of absorbing seed gas molecules to the total pressure in the reservoir, and N is the total number density.

In using fluorescence to measure density variations in a compressible flow one would like all factors multiplying N in equation (1) to be constant. The variation in fluorescent signal would then be directly proportional to the variation in number density. However, in a compressible flow the pressure and temperature vary as well as density and the dependence of equation (1) on these thermodynamic variables must be considered. There are four quantities in equation (1) that depend on pressure and temperature. The quenching rate, Q , and the collision width, $\Delta\nu_c$, are proportional to the collision frequency and each can be expressed as a product of a cross-section, a number density of collision partners, and the average molecular

speed⁹. Therefore, these two quantities have the same pressure and temperature dependence

$$Q = C_q \frac{p}{p_o} \left(\frac{T_o}{T} \right)^{1/2} \quad (4)$$

$$\Delta\nu_c = C_b \frac{p}{p_o} \left(\frac{T_o}{T} \right)^{1/2}, \quad (5)$$

where C_q and C_b are reference collisional quenching and broadening constants respectively, and p_o and T_o are the reference pressure and temperature. The Doppler width, $\Delta\nu_d$, varies as the square root of temperature and the population fraction, f_1 , varies with temperature according to the Boltzmann distribution at thermal equilibrium. The fluorescent signal is, therefore, strongly dependent on pressure and temperature and not simply proportional to density.

We have found that by detuning the laser from the center frequency of the transition this dependence on pressure and temperature can be greatly reduced. In the limit of large detuning, $D \gg 1$, the Voigt function simplifies and equation (1) becomes

$$S_F = C \frac{1}{A_{21} + Q} \frac{\Delta\nu_c}{4\Delta\nu^2 + \Delta\nu_c^2} f_1 N, \quad (6)$$

where the constant

$$C = \eta h\nu \frac{\Omega}{2\pi^2} V_c A_{21} \frac{I}{c} B_{12} f_e$$

is independent of pressure and temperature. In most compressible flow situations the quenching rate greatly exceeds the spontaneous emission rate, $Q \gg A_{21}$, and as long as the laser detuning is large compared to the collision width, $\Delta\nu \gg \Delta\nu_c/2$, equation (6) reduces to

$$S_F = C \frac{C_b}{C_q} \frac{1}{4\Delta\nu^2} f_1 N. \quad (7)$$

Detuning the laser in order to excite the fluorescence off-resonance produces a cancellation of the pressure and temperature dependence of the quenching rate by that of the collision width. Thus, the factors multiplying N in equation (7) are independent of pressure and depend on temperature only through the population factor, f_1 , a dependence which can be made negligible by proper choice of the absorbing transition. The ORLIF signal is directly proportional to the number density, N , and is unaffected by pressure and temperature changes in a compressible flow provided the frequency detuning, $\Delta\nu$, is sufficiently large. Cancellation of the effect of quenching is obtained, however, at the expense of signal strength since large detuning is needed and S_F varies inversely with $\Delta\nu^2$ in equation (7).

Experimental studies were carried out to verify the predicted pressure and temperature independence of the ORLIF signal and to determine the required frequency detuning and resultant reduction of signal strength. Iodine was selected as the seed molecule due to its convenient visible absorption spectrum and ease of insertion into a flowfield. Fluorescence was excited with the 514.5 nm line of the argon-ion laser whose output was frequency tuned, using an intracavity etalon, relative to a strong iodine transition. A preliminary experiment was conducted in a room temperature static cell containing a fixed iodine concentration and variable nitrogen pressure to compare the pressure dependence of the fluorescent signal with that predicted by the theoretical model. Figure 1 shows the variation in the iodine fluorescent signal with nitrogen pressure for four fixed laser detunings from the iodine line center. Iodine transitions adjacent to the primary absorption line limited the available detuning in this experiment to 3 GHz. However, the effect of quenching was significantly reduced by a detuning of only 3 GHz and the resulting decrease in signal level was only a factor of two at one-half atmosphere nitrogen pressure. The theoretical curves shown include the contributions from the adjacent transitions which become significant at a detuning of 3 GHz. The good agreement over the

range of pressure and laser detunings studied confirms the validity of the theoretical prediction.

The underexpanded jet flowfield issuing from a sonic nozzle was chosen as the compressible flow in which to evaluate the ORLIF technique. This flow has a large variation in the thermodynamic variables and it is relatively well understood. Iodine crystals were used to seed nitrogen in a room-temperature reservoir at a pressure of 1137 torr.³ The iodine vapor pressure at room temperature is 0.3 torr, providing a seeding ratio, f_s , of 2.6×10^{-4} . The flow was expanded through a 1.5 mm diameter stainless steel nozzle into a low-pressure chamber to produce the familiar barrel shock structure with a Mach disc located about 4 mm from the nozzle exit.

Results comparing resonance versus off-resonance fluorescence are best displayed by photographs of the fluorescence distribution, in a cross-sectional plane of the jet flowfield, produced by a thin sheet of laser radiation. Figure 2 shows two photographs of the fluorescing flowfield recorded on ASA 400 film. In the top photograph the laser is tuned to the iodine line center, $\Delta\nu = 0$ GHz, and the fluorescence is seen to increase with distance from the nozzle and decrease abruptly across the Mach disc. The density, however, decreases from its sonic value at the nozzle to 3 % of sonic value and then increases by a factor of 4.8 across the normal shock. This photograph clearly illustrates that in resonant LIF the proportionality between fluorescent signal and density in a compressible flowfield is upset by quenching. The bottom photograph shows the effect on the fluorescent distribution of detuning the laser by 3 GHz. The ORLIF signal is seen to decrease with distance from the nozzle through the isentropic expansion and then increase across the shock, as does the density. Data collected pointwise along the axis established that the ORLIF signal was proportional to the calculated number density to within 4 % throughout the isentropic expansion except in the initial region within one-half nozzle diameter of the exit. In this high pressure region of the flow the signal was

not proportional to density since the collision width in equation (6) was not small compared to the maximum available detuning of 3 GHz for the iodine transition used.

With a diagnostic technique available for measuring density one can explore many phenomena of interest in gas dynamics. Figure 3 is an ORLIF photograph of the density distribution in a cross-sectional plane of the same jet flow impinging on a circular cylinder placed where the local Mach number is 4. The dark region above the cylinder is a shadow cast by the cylinder in the laser light sheet. The complex interaction between the underexpanded jet and the cylinder, producing a very low density wake with well defined streams to either side, is quite interesting. This example illustrates the value of the ORLIF technique for quantitative visualization of density in a plane as compared to the standard shadowgraph and schlieren techniques which simply provide information about density derivatives, integrated along the optical path.

The use of more isolated iodine transitions that allow larger detunings and are accessible with tunable, narrow bandwidth visible lasers should allow quantitative density measurements at higher pressures than possible with the argon-ion laser source. Once suitable UV laser sources are available, the application of ORLIF to oxygen should allow density measurements in air without the need for seeding. The approach could also be equally important in the measurement of specie concentrations in chemically-reacting flows where quenching is a major problem when using LIF.

This research was supported by National Aeronautics and Space Administration grants NAG 2-36 and NCC 2-50 and by Air Force Office of Scientific Research contract F49620-80-0091.

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Fig.1. Iodine fluorescent signal versus static cell pressure for four values of laser detuning. Theory given by solid curves.

Fig.2. Photographs of a cross-section of an underexpanded nitrogen jet, seeded with iodine, using resonant (top) and off-resonant (bottom) laser-induced fluorescence.

Fig.3. Photograph of the density distribution in a plane of an underexpanded jet impinging on a circular cylinder, using off-resonant laser-induced iodine fluorescence.

Figure 1

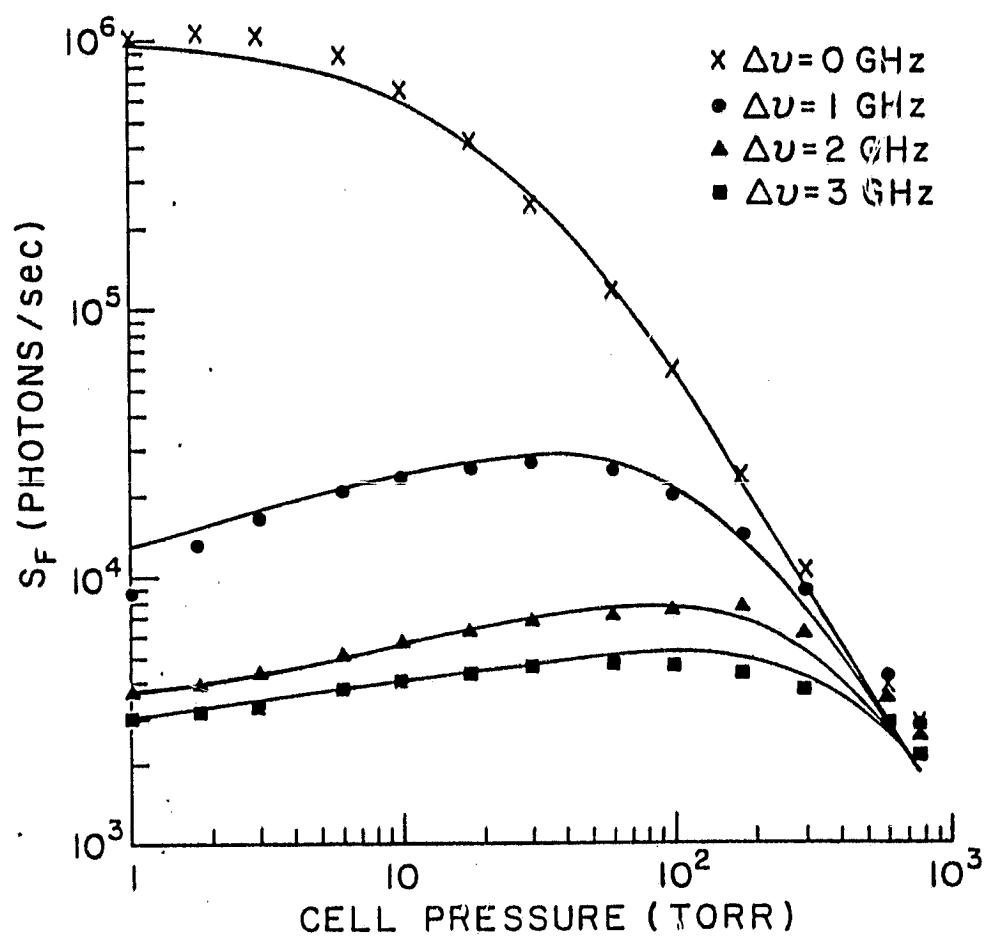


Figure 2

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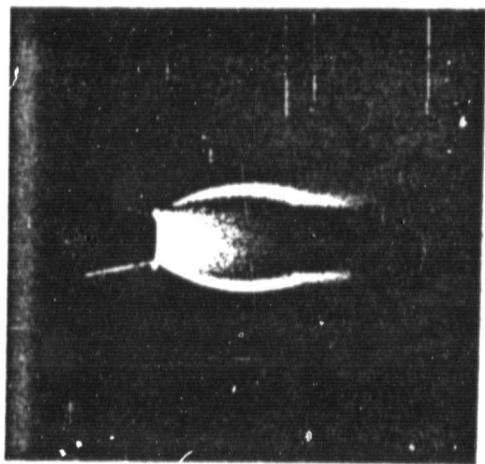
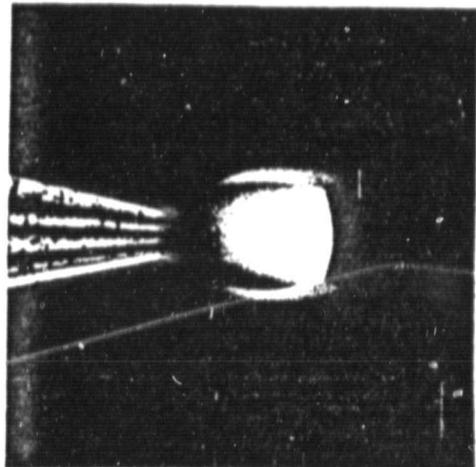


Figure 3

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